LBA Airborne Regional Source Experiment (LARS) and

Transport and Chemistry near the Equator- Brazil (TRACE-B)

An integrated, multi-scale field experiment using aircraft, ground based measurements, and satellites to study the influence of tropical South America on global atmospheric composition and climate, including:

- net sources of CO₂, CH₄, and other radiatively important gases and aerosols
- sources, transformations and export of species determining ozone and the oxidizing power of the atmosphere
- controls on water vapor, aerosols, radicals, NO_x , hydrocarbons, and ozone concentrations in the tropical upper troposphere and lower stratosphere

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1	Motivating Scientific Questions and Goals of the LARS/TRACE-B Mission	2	
	1.1 SCIENTIFIC QUESTION		3
	1.1 Hypotheses		
	1.1 Objectives		5
1	Mission concept 7		
	1.1 THE SCIENTIFIC PROBLEM		7
	1.1 THE APPROACH		
	Table 1: Candidate aircraft measurements for LARS/TRACE-B		8
	Table 2a. Potential aircraft platforms for LARS/TRACE-B in 2003		9
	Table 2b. Measurements on the ground and from space for LARS/TRACE-B		9
1	How the proposed mission will test the hypotheses 11		
	1.1 THE PRINCIPAL FOCI		11
	1.1 A NEW OBSERVATIONAL PARADIGM FOR THIS MISSION		12
	1.1 THE ANALYTICAL AND CONCEPTUAL FRAMEWORK		13
2	Deliverables 15		
3	Linkages 16		
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1 Motivating Scientific Questions and Goals of the LARS/TRACE-B Mission

The global atmospheric abundances of long-lived, radiatively active gases ("greenhouse gases") increased rapidly in the 20th century, reaching concentrations unprecedented in the past million years (or more) of geologic time. These changes have stirred intense scientific interest and public concern. Global increases in concentrations of greenhouse gases and aerosols provide the drive for anthropogenic changes in climate that may have already occurred or that may occur in the future. Increases of photochemical oxidants provide the drive for anthropogenic changes in atmospheric chemistry. Hence very high priorities for atmospheric chemistry are to:

- define rates of change for these and related species,
- develop quantitative understanding of the factors that regulate these changes, and
- provide information to help guide wise societal decisions on management of the global environment to reduce anthropogenic perturbations to atmospheric composition.

The most important radiatively active gases are H₂O, CO₂, CH₄, O₃, N₂O, and CFCs. Aerosols and clouds also play key roles in regulating climate, and both may be significantly affected by human activities. Sources and sinks of greenhouse gases and aerosols are in general qualitatively understood, and the likely factors that underlie recent trends have been identified. However, <u>quantitative</u> understanding of long-term trends is lacking. A major impediment to progress is the lack of data defining regional- and continental-scale sources and sinks, especially in the tropics. We currently lack observational paradigms for measuring net fluxes on continental scales. Since we cannot currently measure net fluxes from the continents to the atmosphere, we cannot detect changes in emission rates, or even define what are the key venues to study.

The mission outlined below is designed to overcome this impediment by developing a new observational paradigm that links observations of atmospheric composition on multiple temporal and spatial scales, simulations of atmospheric transport and chemistry using state-ofthe-art data assimilation models, and measurements from space. It targets the critical scientific hypotheses identified in the Snowmass Planning Workshop for Integration of Satellite Calibration/Validation and Research-Oriented Field Missions in the Next Decade (Snowmass, CO, August 23-27, 1999). It provides an outstanding opportunity for validation of a wide range of satellite measurements from EOS and other platforms in a compelling scientific context. It emphasizes major atmospheric research issues and also includes strong linkages to other NASA programs in the Earth Science Enterprise (Terrestrial Ecology, Land Cover/Land Use Change, Hydrological and Energy Cycles). Brazilian scientists have indicated strong interest and intentions to participate. The mission concept was endorsed by the Steering Committee of the Large-Scale Biosphere-Atmosphere Experiment in Amazônia (LBA) in June 1999. The mission plan has been designed to provide leverage and synergy between its parts as it focuses on the challenging problem of determining fluxes and exchanges on regional and continental scales.

1.1 Scientific question

The LBA Airborne Regional Source Experiment and Transport and Chemistry near the Equator- Brazil (LARS/TRACE-B) mission proposed here represents an integrated biogeochemistry and atmospheric chemistry experiment addressing this scientific question:

• What are the quantitative contributions of the Amazon Basin to the global atmospheric budgets of greenhouse gases, aerosols, oxidants, and their chemical precursors?

What physical, chemical and biological processes regulate these contributions? What are the related implications of rapid development and exploitation of natural resources in and surrounding Amazônia?

This question emphasizes the importance of scale connection from regional processes to global budgets and effects. At this critical transfer of scales, scientific understanding is inadequate to help guide wise policy decisions in areas as diverse as forestry or emissions of pollutants. Amazônia contains the largest expanse of native moist tropical forest in the world. Changes in the carbon stored in these vast forests could significantly influence atmospheric CO₂. The varied mosaic of forests and wetlands releases vast quantities of biogenic gases and aerosols. Vast amounts of combustion products are emitted during the burning season. Deep convection carries these species plus water into the upper tropical troposphere, a region that plays a central role in the Earth's climate. Quantitative estimates of all these fluxes vary so widely among scientists as to prevent credible assessments of future changes. LARS/TRACE-B cuts across the traditional Earth science boundaries of biogeochemistry, atmospheric chemistry, atmospheric dynamics, and radiation to study the tropical atmosphere with an integrated perspective. The mission is strongly coupled to LBA, an ongoing US-Brazilian ground-based program to understand budgets of carbon, energy, and water vapor in Amazônia.

1.2 Hypotheses

The scientific question at the heart of the LARS/TRACE-B mission will address several critical hypotheses identified in the Planning Workshop for Integration of Satellite Calibration/Validation and Research-Oriented Field Missions in the Next Decade (Snowmass, CO, August 23-27, 1999; http://hyperion.gfsc.nasa.gov/Personnel/people/Kawa,_Randy/snow.html). It will test the following hypotheses and sub-hypotheses identified in Tropospheric Chemistry and Dynamics: greenhouse gases, photochemical oxidants, aerosols, nutrients, and global change (Part C of the Snowmass Report)

- Hypothesis #1: Concentrations of long-lived greenhouse gases will continue to rise.
- Hypothesis #2: Changes in anthropogenic emissions and changes in climate will affect atmospheric oxidant concentrations on a global scale.

• Hypothesis #3: Changes in anthropogenic emissions and climate will affect aerosol concentrations on a global scale.

Sub-hypothesis: There is a source of CO_2 of ~1 Gton C/yr due to tropical land clearing. Considerable uncertainty attaches to the very large net emissions of CO_2 attributed to tropical regions, especially due to deforestation and agricultural development. There is also considerable uncertainty regarding carbon emissions from the repeated burning of cerrado and cleared forest areas, which have no long-term effect on CO_2 but are a large source to the atmosphere of other forms of carbon (organic, CO). Recent tower studies have suggested significant net uptake of CO_2 by both "undisturbed" and secondary tropical forests, and cast doubt on the large CO_2 efflux attributed to decay of organic matter in soils after conversion to agricultural use. According to this sub-hypothesis, significant excess CO_2 should be present in the column over Amazônia, after averaging out spatial and temporal variations.

Sub-hypothesis: Changes in land use and in climate in the tropics will modify the structure and function of tropical ecosystems with major impact on the global carbon cycle and consequences for atmospheric CO_2 , CH_4 , N_2O , NO_x , CO, and O_3 . According to this sub-hypothesis, regional emissions that we infer from concentration gradients over developed agricultural areas (e.g. Rondônia) should be measurably different from those over comparable undeveloped areas.

Sub-hypothesis: Changes in anthropogenic emissions of NO_x , hydrocarbons, and CO will affect the abundance of the hydroxyl radical OH, the main atmospheric oxidant. According to this sub-hypothesis, changes in emissions associated with forest clearing, agriculture, biomass burning, and colonization in Amazônia would have a major effect on OH concentrations. The test will involve measurements of OH together with related species along land-use gradients.

Sub-hypothesis: Changes in anthropogenic emissions of NO_x , CO, and hydrocarbons will affect ozone concentrations at all levels of the troposphere, with implications for climate. Better knowledge of NO_x emissions from tropical agriculture and biomass burning, and of NO_x photochemistry in the tropical atmosphere, is important for testing this hypothesis. Current uncertainties in biomass burning emission inventories are particularly severe. The coupling of chemistry and transport over a wide range of scales, including the role of boundary layer dynamics and convection, will be investigated. Better understanding of the natural lightning source of NO_x will be achieved by aircraft measurements in convectively processed air together with analyses of satellite data.

Sub-hypothesis: Climatic perturbations will affect aerosol abundances through changes in biogenic emissions, desertification, and changes in precipitation patterns; complicated aerosol chemistry- climate interactions will result. Tropical ecosystems such as Amazônia represent major sources of organic aerosols to the global atmosphere. The mission will allow an assessment of the sources and fluxes of these and other aerosols, both in the lower and in the upper troposphere where aerosol compositions are expected to be vastly different, and provide a preliminary measure of the sensitivity of the sources to changes in land use. The optical properties of the aerosols will be characterized, including the contrast between the biomass burning aerosols that dominate during the dry season and the natural organic aerosols that

prevail in the wet season. These optical properties will be related to radiative measurements from aircraft and from satellites.

In addition, the program is intended to contribute to the testing of hypotheses on **Water Vapor in the Upper Troposphere and Lower Stratosphere** (Part A of Snowmass report):

- Hypothesis #1: The mixing ratio of water entering the stratosphere in the tropics is set by the average saturation mixing ratio derived from cold-point temperatures between 10° S and 10° N.
- Hypothesis 2: Most of the air entering the stratosphere passes through cirrus clouds in the vicinity of the tropopause.

The ER-2 component of the mission will provide the first regional surveys of $\rm H_2O$ concentrations and temperatures in the cold upper troposphere of this region, the first data on the abundance and distribution of sub-visual cirrus in the region, and a characterization of deep convective outflow. These are the fundamental data needed to design future experiments to test the above hypotheses.

1.3 Objectives

A set of airborne observations is proposed, in concert with long-term measurements and model development ongoing in LBA and with space-borne observations, to measure **regional-scale net exchanges in Amazônia** for environmentally important gases and aerosols. The program of measurements will address:

- <u>The biogenic carbon cycle</u>: measure regional-scale net sources/sinks for: CO₂, ¹³CO₂, ¹⁴CO₂, volatile organic carbon, carbonaceous aerosols, and CH₄, as well as changes in the O₂/N₂ ratio.
- <u>The combustion-derived components of the carbon cycle:</u> quantify the emissions of CO, CO₂, hydrocarbons, nitrogen compounds, soot, and other species due to biomass burning.
- <u>The nitrogen cycle</u>: measure net sources for N_2O , NO_x , NH_3 , and organic nitrogen.
- <u>Atmospheric chemistry</u>: measure sources, transformations and export of reactive gases and aerosols on regional scales including: O₃, NO_y, CO, peroxides, non-methane hydrocarbons (NMHCs), oxygenated organics, sulfur gases, soot, and biogenic aerosols.
- <u>Atmospheric dynamics</u>: measure H₂O, potential temperature, winds, and fluxes of mass and energy from the surface to the mid- and upper troposphere and lower stratosphere.
- <u>Aerosols and radiation</u>: measure aerosol optical depths and single-scattering albedos together with shortwave radiation fluxes.

The mission will be designed to:

- demonstrate the capability to measure and link net surface sources and sinks of important atmospheric species on scales ranging from a region (~100 km x 100 km) to a continent;
- quantify the contributions of surface sources and sinks of trace gases and aerosols on large

scales for a critical region of the global tropics, Brazilian Amazônia;

• define the processes regulating concentrations of greenhouse gases, oxidants, aerosols and their precursors in the atmospheric column over Amazônia and including the climate-critical region of the tropical tropopause.

Aircraft missions necessarily observe the atmosphere for limited time over constrained geographic areas. Altitudes from the surface to the lower stratosphere must be included in an integrated experimental design, because all altitudes are coupled to the surface in an upwelling region such as Amazônia. In order to test the hypotheses, the airborne measurements will be linked downscale to observations at ecosystem level (LBA), to obtain mechanistic understanding of the processes that control net fluxes of greenhouse gases and to define the responses to external forcing such as climate change and management. The airborne measurements will be closely coupled upscale to satellite observations of global and hemispheric distributions, to enable definitive model-based analysis relating observations to fluxes. Satellite data provide the basis for generalizing results of a field campaign to larger scales. We view the multi-scale approach proposed below as essential for a mission intended to address long-term, regional and global issues.

The mission provides an ideal opportunity to validate satellite observations of atmospheric gases and aerosols on the EOS Terra and Aura platforms as well as on other platforms (including the European ENVISAT satellite). A limited listing of these satellite observations is given in Table 2. Of particular importance are the validation of CO from MOPITT; aerosol properties from MODIS and MISR; vertically resolved CO, ozone, NO, and HNO₃ from TES; UT/LS ozone, NO₂, and aerosols from SAGE-3; and HCHO, NO₂ and ozone from GOME and from SCIAMACHY on ENVISAT. A specific advantage of the Amazônia region for validation of satellite observations is the strong convective activity combined with relatively weak horizontal winds, resulting in considerable continental influence and structure over the depth of the tropospheric column. Integration of satellite validation in the LARS/TRACE-B scientific plan is a compelling priority in view of the central role that satellite measurements will play in expanding the spatial and temporal scales of the in situ observations and hence in improving the constraints on the large-scale 3-D models used for data interpretation. The needs of aircraft data for horizontal generalization and the needs of satellite retrievals for vertical definition will thus be addressed in a highly synergistic manner.

We expect also to provide critical tests of an airborne simulator for new technology under development for satellite observations of CO_2 total column. The instrument is a laser-sounding device that ratios atmospheric absorption by CO_2 to absorption by O_2 , in the column of air between the sensor and the ground. If available for this mission, we expect this device to materially aid in the overall objectives by providing for the first time the capability to measure total CO_2 column amounts, day or night. Validation with the in situ measurements of CO_2 would provide a major step in developing a space-borne capability for CO_2 flux measurements from space.

The mission will be a component of LBA, a major international collaborative study led by Brazil with strong United States involvement. The goal of LBA is to improve knowledge of the moisture, energy, biogeochemical, aerosol, and trace gas budgets in the Amazon Basin and their perturbation by human activity. The mission will complement and enhance this activity. The LBA ground-based, ecological, and meteorological experiments provide a foundation and leverage for the mission as well as an effective framework for developing collaborations with atmospheric chemists from Brazil.

<u>2</u> Mission concept

2.1 The Scientific Problem

LARS/TRACE-B will deploy a coordinated set of research aircraft in concert with satellites and LBA ground sites to determine regional sources and sinks by direct measurement. The suite of aircraft must have operational capabilities from the Planetary Boundary Layer (PBL) to the Upper Troposphere/Lower Stratosphere (UT/LS). A comprehensive ensemble of observations will be undertaken, set in a framework of conceptual and detailed models, to provide robust regional source estimates for the most important gases and aerosols.

2.2 The Approach

The aircraft will be deployed to obtain temporal and spatial integration of atmospheric profiles of gases, aerosols, and radiation. Two deployments are envisioned: one in the wet season or at the transition from wet to dry, and one in the dry/burning season. A comprehensive suite of tracers will be measured and linked to meteorological data and models, to long-term ground-based data from LBA, and to remote sensing data. Low altitudes are generally regions of atmospheric inflow, since there is strong convergence over Amazônia on average; data will be obtained using **ground stations** and **low-flying aircraft**. At high altitudes, regions of inflow, outflow, and injection by deep convection, and the near-tropopause region, will be sampled using **high-flying aircraft** and **satellites**. Deep convective storms occur regularly throughout the region during the wet season, and remain prevalent in parts of Amazônia in the dry season. Thus all levels of the atmosphere must be considered to be in direct communication with the surface, and the atmospheric concentrations of gases and aerosols should be measured from the surface to the tropopause to achieve the goals of the mission.

We envision the following aircraft for implementing this strategy:

- 1. Lowest altitudes (150 8,000 m): **INPE Bandeirante** and/or **UND Citation II**, or equivalent.
- 2. Low to Middle altitudes (300 8,000 m): NCAR C-130 or NASA P-3.
- 3. Middle and Upper altitudes (10,000-12,000 m), and long-range outflow: NASA DC-8.
- 4. Upper Troposphere and Lower Stratosphere (12,000 17,000 m): NASA ER-2.

Table 1 presents a possible list of measurements on the airborne platforms, with preliminary assignment of priorities, and Table 2 lists the ensemble of platforms expected to be deployed during the mission. Table 2b identifies the suite of important satellite instruments for addressing the goals of the mission. It is expected that the validation component of the mission will focus on these instruments.

Table 1: Candidate aircraft measurements for LARS/TRACE-B

Codes: 1 = mission-critical, 2 = very important, 3 = important, () = improvement or validation of current instrumentation between now and flight time is necessary. †Under development; priority ranking *if available*. Tabulated detection limits and time resolutions are minimum requirements.

Species/Parameter	Priority	Detection limit	Time resolution
CO_2	1	0.1 ppmv (precision)	1 s
CH ₄	1	20 ppbv (precision)	1 s
N_2O	1	0.5 ppbv (precision)	1 s
H_2O	1	3 ppmv or 1%	1 s
O ₃ (in situ)	1	3 ppbv	1 s
СО	1	5 ppbv	1 s
remote aerosols	1		
remote ozone (nadir/zenith)	1		
eddy-covariance fluxes	1	depends on species	20-30 minutes
volatile organic carbon	1	50 pptC	< 1 minute
compounds			
remote total column CO ₂ or	2†	< 1ppm variations in	1 minute
CO ₂ LIDAR †		space and time	
O /NI	2	2.2	.1
O ₂ /N ₂	2	2-3 per meg	< 1 minute
NO	2	3 ppt	1 s
halocarbons including methyl halides	2	2 pptv	1 min
OH	2	$5x10^5 \text{ cm}^3$	1 min
HO ₂	2	$1 \times 10^{8} \text{ cm}^{3}$	1 min
NO ₂	2	5 pptv	1 min
HNO ₃	2	30 pptv	2 min
PAN	2	10 pptv	2 min
NO _v	2	50 pptv	1 s
aerosol size distribution	2	$10/\text{cm}^3 (0.3-0.6 \text{ um})$	10 s
aerosol composition	2	10 pmol mol ⁻¹	5 min
ultrafine aerosols	2	3-10 nm	1 min
aerosol optical depth	2	0.2	5 min
aerosol carbon mass	2	50 ng m ⁻³	15 min
aerosol absorption cross-	2	$5x10^{-6}$	
section			15 min
aerosol scattering cross-	2	$5x10^{-5}$	10 min
section			
aerosol backscattering cross-	2	$5x10^{-6}$	10 min
section			
long- and short-wave	2	(spectrally resolved)	
radiation fluxes (up/down)	1		l

COS	2	10 pptv	5 min
²²² Rn	2	5 pCi/SCM	5 min
²¹⁰ Pb	2	1 fCi/SCM	5 min
⁷ Be	2	0.1 mBq/SCM	5 min
UV actinic fluxes	2	0.1 µW nm ⁻¹ cm ⁻²	30 s
storm scope	2	range 400 km	<3 min hold time
НСНО	2	10 pptv	5 min
H ₂ O ₂ , CH ₃ OOH	2	10 pptv	5 min
organic nitrates	3	10 pptv	5 min
DMS	3	1 pptv	5 min
H_2S	3	5 pptv	5 min
SO_2	3	5 pptv	5 min
$H_2SO_4(g)$	3	$2x10^5 \text{ cm}^{-3}$	5 min

Highly capable, proven, well-calibrated instruments and associated science teams are now available for the full complement of aircraft sensors. There is strong interest at NASA, NCAR, U.S. universities, INPE, and other major Brazilian institutions. All of the aircraft noted above have operated in Brazil in the last 5 years. These recent developments allow us to plan a much more comprehensive and rigorous mission than possible hitherto.

Table 2a. Potential aircraft platforms for LARS/TRACE-B in 2003

Aircraft	Vertical range	Measurements	Targeted objectives
Citation or Bandeirante	0.15-8 km	In situ chemistry	Diurnal cycle of PBL dynamics and chemistry, relation to surface fluxes
P-3 or C-130	0.15-8 km	Detailed in situ chemistry and micrometeorology, eddy correlation fluxes, radiation	Biosphere-atmosphere exchange, boundary layer dynamics and chemistry, radiation budget.
DC-8	0.3-12 km	Detailed in situ and remote chemistry	Regional-scale distributions, net export
ER-2	12-20 km	in situ chemistry, radiation	UT/LS: radiation, near- tropopause processes and chemistry, convective outflow, H ₂ O budget

Observations of HCHO, NO₂, and tropospheric ozone from GOME and SCIAMACHY, CO and aerosols from TERRA, and vertically resolved ozone, CO, H₂O, CH₄ and other tracers from TES (Table 2b), are of particular interest for addressing the scientific objectives of LARS/TRACE-B. Validation of these satellite observations using the *in situ* observations from aircraft will be an integral part of the mission execution plan. The validated satellite data will in turn provide important information for the mission science objectives. For example, recent work indicates that the HCHO column measurements from GOME and SCIAMACHY may provide a measure of column rates for oxidation of reactive hydrocarbons, and this quantity would in turn represent a proxy for reactive hydrocarbon emission fluxes. Column amounts of HCHO are expected to be controlled largely by emissions of isoprene. In LARS/TRACE-B

the intention would be to test the accuracy of the column retrievals, and to critically examine emission flux estimates using airborne and ground-based data. Then the satellite data will be used to map isoprene emissions on the scale of Amazônia. Data on NO_y species from SAGE-3, TES, GOME, and SCIAMACHY will provide information on the generation of NO_x from lightning.

Table 2b. Measurements on the ground and from space for LARS/TRACE-B

	Platform	Measurements	LARS/TRACE-B objectives
Ground- based			
	Towers	Detailed chemistry and micrometeorology, eddy correlation fluxes, other flux measurements	Biosphere-atmosphere exchange, local hydrological and radiation budgets
	Ozonesondes	Ozone, T, H ₂ O	Regional oxidant budget, atmospheric structure
	Balloons	Chemistry, H ₂ O	UT/LS processes
Satellites			
	TOMS/TRIANA (1979)	Tropospheric ozone column, aerosol index	Ozone and aerosol budgets, biomass burning
	GOME (1995)	Tropospheric ozone, HCHO, NO ₂ , aerosol columns	Ozone and aerosol budgets, reactive hydrocarbon emissions, lightning NO _x , photochemistry
	OTD (1995)	Lightning Discharge	Lightning NO _x production
	MOPITT (1999)	CO, methane column	Biomass burning, methane budget
	MODIS/MISR (1999)	Aerosol optical depth, Ångstrom coefficient, spectral radiances, other parameters.	Aerosol abundances, atmospheric transport, role of aerosols in radiative balance
	SCIAMACHY (2001)	Tropospheric ozone, HCHO, NO ₂ columns, concentrations in upper troposphere	Same as GOME + UT/LS chemistry, stratospheric age of air
	MIPAS (2001)	Tropospheric ozone, CO, H ₂ O (limb)	Convective outflow, tropopause processes
	SAGE-3 (2003)	UT/LS ozone, NO ₂ , aerosols	UT/LS dynamics and chemistry
	TES (2003)	Tropospheric ozone, CO, H ₂ O; UT/LS NO, HNO ₃	Tropospheric dynamics and chemistry, regional chemical budgets, lightning NO _x , UT/LS processes
	HIRDLS (2003)	UT/LS ozone, H ₂ O, HNO ₃	UT/LS processes
	MLS (2003)	UT/LS H ₂ O, ozone, HCN	Convective outflow, UT/LS processes
	OMI (2003)	Tropospheric ozone, HCHO, NO ₂ , aerosol columns	Same as GOME

Most flights will be over the Brazilian Amazon region, from Brasilia north, except that the DC-8 and ER-2 will conduct flights in the periphery including over the oceans.. Low-altitude and airborne eddy-flux flights will leverage from the ground stations established under LBA, as

shown in Figure 1 (beige rectangles).



Figure 1. Map of South America with Brazil highlighted (yellow), and major ground stations of the LBA project indicated as beige rectangles.

3 How the proposed mission will test the hypotheses

3.1 The principal foci

The foci of tests of the principal hypotheses will be to quantify and understand the sources and sinks at the earth's surface of greenhouse gases, aerosol-forming gases, and primary pollutants, at spatial scales from regional to continental. The associated atmospheric chemical reactions and production and loss of O₃ and radicals will also be studied.

To test the principal hypotheses and sub-hypotheses identified in the Snowmass report and laid out in section 1.2, we need to know the "big picture" about surface sources and sinks. What are the fluxes and their variations in time and space, over large areas? Is the ocean or the land dominant, what are the magnitudes of the biological sources and the combustion sources? Are changes in land use and climate affecting these sources or sinks significantly and thus changing the global concentrations? The elements that factor into the big picture include fossil fuel and biomass burning, land management, agriculture, ecosystem processes, weather, and physical processes such as flooding or erosion.

The proposed mission tackles critical aspects of the Snowmass hypotheses by measuring the

net sources or sinks of key gases and aerosols in Amazônia. The influence of the undisturbed tropical forest will be determined, and the perturbations associated with forest clearing and conversion to agriculture will be elucidated. The experimental design covers multiple temporal and spatial scales in order to provide this information in the *global* context. The experiment design itself represents a test of a key, albeit implicit, hypothesis, viz. *it is possible to combine atmospheric observations with regional and global data-assimilation models to determine regional and continental net fluxes to/from the atmosphere quantitatively.* The extension of the observations to the tropical UT/LS provides an excellent opportunity to leverage radiative flux measurements onto the observations of chemistry, including the effects of sub-visual cirrus clouds, addressing a subset of the water vapor/climate hypotheses.

A critical focus for testing each hypothesis will be an effort to prove the associated **null hypotheses**. Efforts to prove each **null hypothesis** will be included as an essential component of the experiment design. A useful example is provided by CO₂ over Amazônia. Global analyses indicate a large regional net source from deforestation, but tower flux data have been reported showing large uptake. The LARS/TRACE-B observations could be very effective in proving a null hypothesis. For example the data might show the absence of the excess CO₂ expected in association with the CO from biomass burning, or the lack of the draw-down of CO₂ column concentrations anticipated if large biogenic sinks are active. Likewise, agricultural development is expected to lead to enhanced outputs of many photochemical precursors. A null hypothesis would be proven if concentrations of these gases did not show the expected elevated levels over areas of Amazônia currently developed for agriculture.

3.2 A new observational paradigm for this mission

The effort to *quantify and understand sources and sinks of greenhouse gases, aerosol-forming gases, aerosols, and oxidant precursors in the atmosphere* requires new observational paradigms. The attempt to get the "big picture" brings us to the frontier of research. Observations have rarely (never?) been undertaken that combine measurements on the relevant range of spatial and temporal scales to obtain, quantitatively and convincingly, regional scale net fluxes. One obvious motivation to link atmospheric chemistry with studies of CO₂ and other long-lived greenhouse gases is the strong purchase provided by studying multiple tracers at once. Each tracer or class of tracers provides independent information to constrain processes and rates of emission and deposition. Moreover, CO₂ is the primary metabolite of ecosystems, and CO and CO₂ are the principal effluents of combustion, hence concentration gradients for these gases commonly exhibit strong covariance with other species produced by metabolic and combustion processes. Quantitative budgets and fluxes for CO and CO₂ provide the key information needed to assess net fluxes for many other species.

3.3 The analytical and conceptual framework

A. Measure atmospheric distributions of species concentrations over Amazônia and around its periphery, and analyze the data in terms of *continental-scale fluxes* using regional and global data assimilation models.

Concentrations of each species will be measured at low altitude over the full diurnal cycle, using the INPE Bandeirante and/or UND Citation II (or equivalent), exploiting the flexibility for these platforms to make extensive measurements in the planetary boundary layer. Concentrations in the nocturnal boundary layer (too low for flight) will be measured by LIDAR, if the planned CO₂ laser sounder comes on line, and by towers and tethersondes. Observations by aircraft will be placed in the context of continuous long-term measurements of concentrations and fluxes at LBA towers and at the long-term monitoring site of the LBA project at the critical inflow region, on the East Coast near Natal.

From the lowest aircraft altitude to middle troposphere, measurements will use the NCAR C-130 or the NASA P-3, exploiting the capability for low-and mid-altitude flight, eddy-covariance flux measurements, and long range.

To cover long distances to characterize advective import from, and export to, the global atmosphere, to determine the influence of convective outflows up to 12 km altitude, and to deploy remote-sensing DIAL systems, the DC-8 is the preferred platform. In the upper troposphere and lower stratosphere we plan to deploy the ER-2 to obtain first-time data on concentrations and radiative fluxes in this critical region, including the role of deep convection and lightning on NO_x and other species.

Analyses of these data will be undertaken by both conventional, forward-modeling approaches, in which sources and sinks are specified, concentration distributions computed, and comparisons made to the observations, and by newer inverse models using a range of state-of-the-art analytical strategies. We envisage a suite of models from regional to global scale using assimilated meteorological data for the period of observation.

From part A we will obtain regional and continental scale concentration gradients for key species, with generalized net flux estimates obtained via the atmospheric models.

B. Obtain regional fluxes using a suite of independent analysis methods.

1. <u>Column average anomaly method.</u> Analyze the differences between the column average concentrations above and below the highest altitude reached by the PBL, for a comprehensive ensemble of locations and times (time-dependent column amount method):

Net Exch. =
$$-\left(\left\langle q_{b}\right\rangle - \left\langle q_{h}\right\rangle\right) n_{h} h / \tau_{r}$$

[q = mixing ratio, h = maximum altitude reached by the PBL, q_b = vertical-mean q from 0 to h, τ_r mean replacement time for the layer, n_h atmospheric density at h, $\langle \rangle$ = observed 24-hr mean]. This approach should be applied for CO, CO₂, H₂O, and other gases with sufficiently long atmospheric residence times to insure that excess amounts observed in a column will be transported to the environment.

The experiment is designed to acquire sufficient *in situ* data to allow accurate determination of column amounts for this purpose. We anticipate an important role also for new airborne remote column measurements of CO₂ or other gases (e.g. CH₄ LIDAR, CO column), currently under development. Current plans suggest that devices may be ready by 2003, and would immediately provide new, potent tools for regional flux assessments.

- 2. Traditional <u>CBL</u> budget analysis using entrainment rates from towers and from the aircraft, with the same data as used in (1).
- 3. Flux- gradient relationships in the "mixed" layer, combining observed concentration profiles with aircraft and tower observations of turbulence quantities and eddy-covariance fluxes to derive fluxes from vertical profiles. This approach may be used for a variety of chemical species and aerosols. These are in effect <u>flux ratio methods</u> in which **direct flux measurements determined by eddy covariance or PBL or CBL budgets** are combined with concentration data to derive fluxes for species not currently available for direct observations of flux.
- 4. <u>Data assimilation models</u> of several types (Eulerian, Lagrangian; regional, global) will be coupled to biome models (e.g. SiB2), using space, aircraft and tower observations to constrain biotic exchanges. Parcel-following, or flow-following studies would fall into this category.

The approaches described in A and B above use a suite of transport models where the objective is to combine assimilated meteorological data and observations of trace gas and aerosol distributions to infer large-scale fluxes directly. The work will extend traditional methods for assessing sources using measurements of concentrations and comparing to limited data by obtaining a sufficient set of measurements to allow robust estimation of net fluxes from observations.

C. Determine aerosol composition, sources, and seasonality in the context of the short-wave energy balance for tropical South America.

<u>1 Aerosol sources.</u> Analyze the fundamental optical properties of the aerosol on a series of generalized boundary-layer flight tracks. Use trajectories, chemical tracers, and regional transport models to determine the origins and transport pathways of aerosol plumes. Relate

observations of biomass burning aerosols to amount and type of combustion using chemical tracers, and compare to carbon burn rate and fire intensity data from satellite observations. This work will build on years of ongoing collaborative research between NASA and Brazilian groups.

<u>2 Effects of aerosols on the radiation budget.</u> Studies will concentrate on linking compositional and optical aerosol observations to integral radiative properties that are deducible from satellite observation.

A requirement for cross-disciplinary studies is evident in the questions being addressed. One cannot directly study all regional-scale source or deposition processes, legacies of land use or current management, or emissions due to burning of particular types of crops. To *understand* the sources, to be able to assess what will happen in other places or at other times, and to follow long-term changes in sources and sinks, studies must proceed on the ecosystem scale and address ecosystem processes in the same locale as the aircraft measurements. A balanced suite of site-specific measurements must be carried out, and a subset should be sustained for periods commensurate with the long-term trends that drive global change. A major objective therefore is to link observations at particular sites unambiguously to airborne and satellite measurements at the larger scales.

4 Deliverables

- (1) Demonstration of a new paradigm for measuring net fluxes to the atmosphere on regional and global scales.
- (2) Determination of the biosphere-atmosphere fluxes of greenhouse gases, oxidants, aerosols, and related species over the range of ecosystems in tropical South America.
- (3) Characterization of the concentrations of greenhouse gases, oxidants, aerosols, and related species in tropical South America.
- (4) Quantitative estimates of the net flows of these species across the boundaries of the Amazon Basin.
- (5) Definition of the structure of the continental boundary layer and relation to both (a) the distribution of atmospheric gases and aerosols exchanging with the surface and (b) the fundamental surface-moisture parameters which determine both emissions and atmospheric mixing.
- (6) Direct measurements of the vertical redistribution of gases and aerosols in the tropospheric column associated with deep convective systems near the Equator.
- (7) Observations defining the factors that control the concentrations of H₂O, ozone, nitrogen oxides, and aerosols in the UT/LS, including the role of Amazônian deep convection.
- (8) Validation of satellite sensors including TOMS, MOPITT, MODIS, MISR, SAGE-3, and the Aura payload (TES, HIRDLS, MLS, OMI), for the suite of species listed in Table 2b.
- (9) Field tests and proof-of-concept of the airborne simulator for space-borne atmospheric measurements of CO₂ columns (N.B., possibly for other gases such as CO), if available as currently planned.

5 Linkages

The LARS/TRACE-B mission has a compelling rationale for investigating key science questions of societal importance, motivated by the *NAS Pathways Report* and the *US Carbon Cycle Science Plan*. It will provide • quantitative data defining the global role of Amazônia in the cycles of CO₂, CH₄, and other gases; • quantitative data defining the global role of Amazônia in the budgets of reactive trace gases and aerosols; • 1st -order tests of analysis of regional budgets using 3-D atmospheric transport and chemistry models; • tests and implementation of new concepts for determining regional-scale fluxes.

Beyond its atmospheric chemistry and physics focus, the mission provides strong linkages to other NASA programs in the Earth Science Enterprise (Terrestrial Ecology, Land Cover/Land Use Change, Hydrological and Energy Cycles) and also programs at NCAR (Climate and Global Dynamics Division, Environmental and Social impacts). The mission will link strongly to ongoing LBA activities focused on dynamical, hydrological, and biogeochemical aspects of the interactions between Amazônia and the global environment.

Specific linkages include:

LBA towers: Towers provide continuous measurements of concentrations and fluxes. These data can anchor airborne and space-borne observations in both space and time. The tower observations provide the framework to extend aircraft data for limited flight days to long-term means. Airborne data in turn provide regional spatial gradients critical for interpreting tower data in the representative regional context.

Data assimilation models: Chemical transport models driven by assimilated meteorological data provide a primary analysis tool for relating atmospheric concentration data to surface boundary conditions (i.e., biosphere-atmosphere exchange fluxes) and to net export fluxes across the boundaries of the region.

Space-based observations: Satellite data place airborne regional-scale observations in the basin-wide context for CO, ozone, H₂O, CH₄, and other species (see Table 2b). Data from LARS/TRACE-B, stretching from the surface to the lower stratosphere, will provide the most comprehensive and rigorous validation data for satellite observations in the tropics. Satellite algorithms are extremely sensitive to errors in regions of high gradients in temperature and concentrations, and from that standpoint the Amazônia region is ideally suited for validation exercises.

Climate models: Radiative fluxes in the upper troposphere over Amazônia play a critical role in the earth's climate. The proposed measurements will provide the first detailed examination

of the factors regulating long-wave and short-wave radiation in this critical region including concentrations of H_2O and ozone, aerosol distributions and optical properties, and sub-visual cirrus clouds.

Airborne CO₂ column and DIAL measurements: ESE has initiated development of active sensors to measure total column CO₂ and to make height-resolved DIAL measurements from space. Other gases, especially CO, might be included. Laboratory studies are currently underway, and success in demonstrating feasibility appears likely. The next step would be to build an airborne simulator. We would expect to deploy this airborne simulator in LARS/TRACE-B, if it is ready as currently planned. Inclusion of this new sensor in the experiment would allow us to provide a comprehensive test of its capabilities. It would also contribute enormously to the goals of the experiment. For example, by allowing us to observe the build-up of CO₂ at night over a large region, down to the canopy top, accurate determination of the rate for nocturnal respiration would be possible. The technique would considerably enhance the application of time variations in the total column to determine the regional net source or sink for CO₂.